Metallic state in $(TMTSF)_2PF_6$ at low pressure

I. J. Lee* and M. J. Naughton†
Department of Physics, State University of New York, Buffalo, New York 14260-1500
(Received 11 September 1998)

Magnetoresistance rotation data in the pressure range 6.0–8.3 kbar in the quasi-one-dimensional metal $(TMTSF)_2PF_6$ are well reproduced by semiclassical simulations. Thus, benchmarks suggested by an incoherent interlayer transport model for non-Fermi-liquid behavior are not met, in contrast to results at higher pressures. A saturating magnetoresistance, however, appears nonclassical. More definitive criteria on what is and is not a Fermi liquid are required before the ultimate fate of this and related low-dimensional conductors can be determined. [S0163-1829(98)51244-1]

The molecular conductors $(TMTSF)_2X$ are low-dimensional materials with simple band structures yet rich electronic properties, from superconducting to magnetic insulating. When a magnetic field is rotated in the y-z plane [using $(a,b,c)=(x,y,z)$], the field components along y and z produce new periodicities in the electron motion, defined by $\omega_y=ebv_FH\cos\theta/\hbar$ and $\omega_z=ecv_FH\sin\theta/\hbar$, where v_F is the Fermi velocity, b and c are lattice spacings, and ω_i is the frequency with which the electron crosses the Brillouin zone in direction i.[1] With the field aligned at an angle where these frequencies are commensurate, such that $(c/b)\tan\theta=p/q$ (p and q are integers), electrons will have a nonzero average velocity along H, enhancing the conductivity. Thus, resistance anomalies are expected,[2] observed,[3] and explained semiclassically[4] at these so-called Lebed magic angles.

An alternate explanation of this magic angle effect was recently suggested by Strong, Clarke, and Anderson (SCA),[5] who argue that an ''inelasticity'' ev_FH_yc is introduced to an electron's motion by an in-plane magnetic-field component along the y direction. When H_y is large enough such that this inelasticity is comparable to the coherent-motion interlayer hopping energy t_z, the system becomes effectively two dimensional by a virtual vanishing of t_z. On the other hand, when the field is aligned along the real-space lattice direction (the usual magic angle configuration), higher order hopping directed along the field survives even if the coherent part of t_z vanishes. Thus, for every arbitrary angle, the system is effectively a two-dimensional non-Fermi liquid (NFL), except near the z axis and the magic angles, where the system maintains its three dimensionality. The system is thus on the verge of a three-dimensional Fermi-liquid state which, in this model, totally hinges on a very small amount of coherent interlayer transport at zero field.

A second angular effect was found in $(TMTSF)_2ClO_4$ when the field was rotated in the crystal x-z plane.[6] The semiclassical Boltzmann equation was successfully applied, and the hopping ratio t_y/t_x was deduced by matching features in the data with points where the electron velocity averages out effectively.[6] A similar x-z resonance, but with very different magnetoresistance background, was found by Danner and Chaikin (DC) in PF_6 at 9.8 kbar.[7] The effect of a small H_y, above which the characteristics of the x-z resonance vanish, was asserted as evidence for the vanishing of the coherent interlayer transport t_z, thereby yielding a NFL as per the SCA scenario. In addition, power-law scaling in z-axis resistance versus the z component of the field was given as additional confirmation of NFL behavior resulting from the SCA incoherence.[7]

In this paper, we present data from simulations and experiments on $(TMTSF)_2PF_6$ in the pressure range 6.0–8.3 kbar that do not exhibit there non-Fermi-liquid features. In magnetic-field rotations from x to z with and without a field component along y, magnetoresistance oscillations very similar to those found in $(TMTSF)_2ClO_4$, but more distinct, were found. Simulations using semiclassical Boltzmann transport reproduce the data very well over the entire experimental range. In the pressure range employed, using the criteria defined in Refs. 5 and 7, we do not find evidence for NFL behavior.

The experiments were performed in a dilution refrigerator in a split coil magnet, with internal and external goniometers providing *in situ* dual axis rotation (ϕ and θ). A BeCu pressure cell[8] provided quasihydrostatic pressure up to 8.3 kbar. The interlayer resistance R_{zz} was measured by the conventional ac technique. R_{zz} for x-z rotations, with various third direction components H_y at 8.3 kbar, 0.32 K, and 7 T, are shown in Fig. 1. The behavior of the background here is quite different from what was observed previously on PF_6 at 9.8 kbar by DC,[7] where the detailed resonance (oscillation) structure was not quite so evident in the raw data. As the tilt angle ϕ increases, the peaks and dips shift away from the x axis ($\theta=0$). For a given tilt ϕ, their positions are independent of H and T, suggesting a Fermi-surface topology origin, as noted previously for ClO_4.[6] Several characteristic features of the x-z rotation, such as the shallow minimum about, and small peak at, the x axis, and the major maxima near $\theta=\pm15°$, remain at least until $\phi=7°$. This is so even with the angular positions of the peaks beyond $\theta=4°$ being shifted by an interplay of this x-z resonance effect with the Lebed y-z resonance, which is responsible for the asymmetry in the data.

If we employ the same argument as DC,[7] we can estimate from our data a lower bound for the characteristic field at which the x-z resonance, and thus the coherent part of t_z, vanishes as $\mu_0H_y^*>0.85$ T (i.e., 7 T sin7°). That is, the x-z effect still remains in this 7° tilt. This is to be compared with

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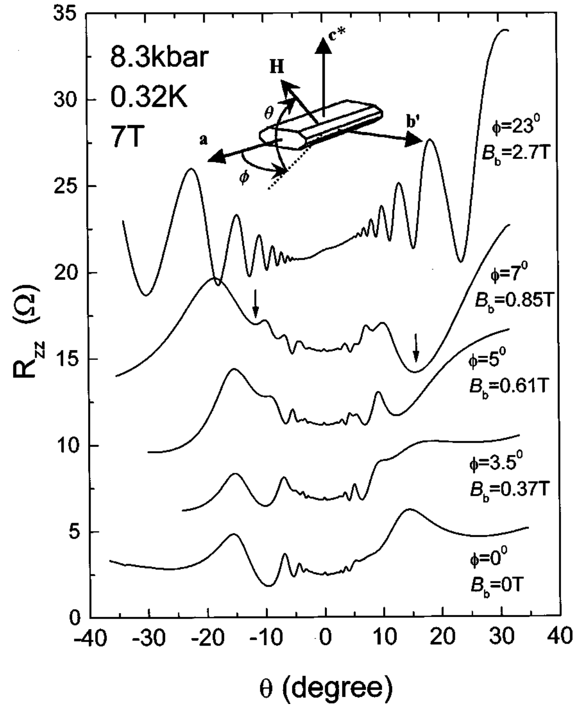

FIG. 1. Interlayer magnetoresistance x-z (a-c) plane rotations with varying tilts (offset) in a 7-T field, showing x-z resonance effect coexisting with y-z magic angle effect. Tilt angles ϕ and field components along y, H_y, are indicated.

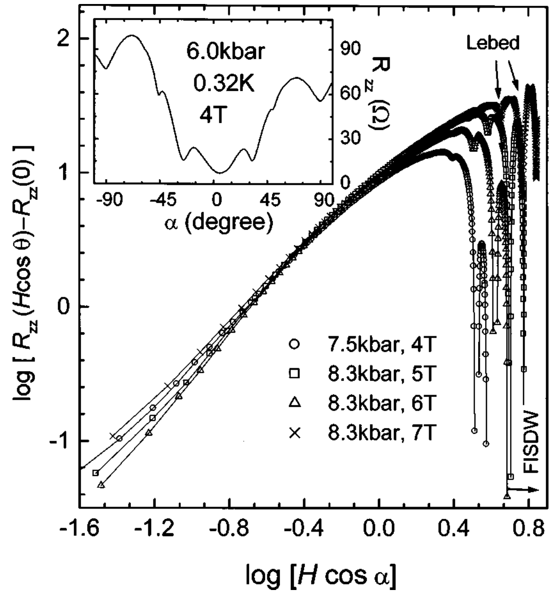

FIG. 2. Scaling of Lebed y-z rotations versus the interlayer field H_z at 7.5 and 8.3 kbar. The slope changes smoothly throughout, and so does not display power-law behavior anticipated by SCA for the non-Fermi-liquid state. The inset shows that at a lower pressure, 6.0 kbar, a plot as in the main panel cannot be made, with $R_{zz} \sim \sin\alpha$ (classical).

the decoupling or confinement field obtained in the DC experiment, $\mu_0 H_y^* = 0.2$ T.[7] In fact, one should expect the coherent part of t_z to *decrease* in this system with decreasing pressure, by an amount proportional to the overlap of the π-electron orbitals. On the contrary, our H_y^* at 8.3 kbar is more than four times *larger* than that obtained previously in the same system at 9.8 kbar.

Enhancement of interchain coupling with pressure in $(TMTSF)_2PF_6$ was firmly established in a study of the pressure dependence of the shortest Se-Se and Se-F interactions by Gallois *et al.*,[9] with a similar observation by Beno *et al.*[10] for a sibling compound, $(TMTSF)_2AsF_6$. For these materials, the trend toward higher dimensionality naturally explains the decrease in the metal-SDW (Peierls) transition temperature, and thus the importance of electron correlations, with pressure. Since NFL behavior should be favored in a system with smaller t_z (in other words, in PF_6 at lower pressure), one would expect in our experiment at 8.3 kbar a vanishing of the x-z resonance almost as soon as H_y is turned on, in contrast to what is observed. Here, the magic angle effect distorts the x-z effect, resulting in a highly asymmetric resonance structure, but the x-z effect nontheless remains, *coexisting* with the magic angle resonance. See, for example, the arrows in Fig. 1 near $\theta = -12°$ (magic angle $p/q = +1$) and $+15°$ ($p/q = -1$) for $\phi = 7°$. Therefore, an important question is whether incoherent interlayer transport as per SCA and DC is responsible for the disappearance of the x-z effect.

A second indicator of NFL behavior suggested by SCA and DC was noninteger power-law behavior in scaled magnetoresistance from y-z plane rotations versus the interlayer field H_z. In Fig. 2, we show magnetoresistance obtained from such Lebed rotations at three pressures. In the main figure are data at 7.5 kbar (○=4 T), and 8.3 kbar (□, △, ×=5,6,7 T). Some scaling may be evident at intermediate $H \cos\alpha$ (α is the angle from the z axis; log=base 10), but power-law behavior is clearly not observed, as the slope continually changes. In addition, the power-law scaling should be most noticeable as the applied field approaches the y axis, on the left side of Fig. 2, where only incoherent interlayer hopping remains, according to SCA. This is not the case here. The features to the right are Lebed resistivity dips. The inset shows a y-z rotation at 6.0 kbar. In this case, the data simply *cannot* be scaled in the manner suggested by SCA, since the resistance along the z axis is a global minimum instead of a maximum. To repeat, a background magnetoresistance scaling as $(H \cos\alpha)^\gamma$ with some noninteger power γ is expected by SCA's version of NFL (via decoupling).[5,7] Instead, as pressure is reduced to 6.0 kbar, the observed behavior (inset) is classical ($\sin\alpha$). So, the conditions for SCA's NFL arguments to hold appear much worse as the applied pressure is reduced, in opposition to our understanding of the anticipated role of pressure.

Next, we compare our experimental results in Fig. 1 with semiclassical calculations appropriate for a quasi-one-dimensional (1D) conductor. Simulations of x-z rotations with various field tilts along y are shown in Fig. 3, where we used an orthorhombic symmetry with lattice constants $a = 6.98$ Å, $b = 7.85$ Å, and $c = 13.26$ Å, as measured in PF_6 at 7 kbar and 1.7 K.[11] Details of the computer simulations are given in Ref. 12, where similar x-y plane calculations were performed. The overall features, such as the angular positions and relative sizes of the peaks, are reproduced quite well in the simulations, again using the simplified Boltzmann transport equation.[12] Notice the near-perfect match between experiment and simulation of the positions of the peaks for the pure x-z rotation shown at the top of Fig. 3. The structure

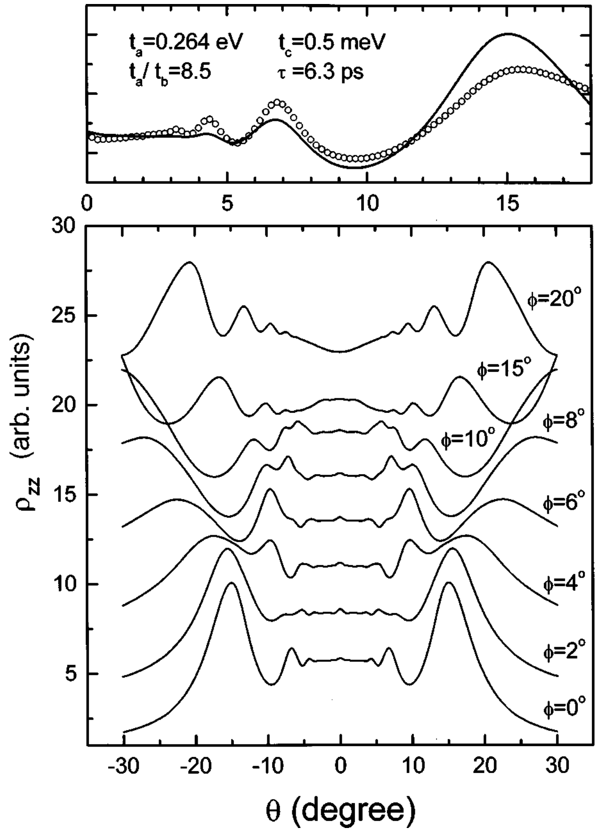

FIG. 3. Calculated resistivity ρ_{zz} versus x-z angle θ for different y-direction tilts ϕ (offset) of a 7-T field, reproducing the evolutions of the x-z and y-z resonance effects of Fig. 1, including their coexistence. Top: Direct comparison of simulation (line) and data (circles) for the $\phi=0$ x-z rotation.

near $\theta=0$ (see also data in Fig. 1) is related to closed orbits and Fermi-surface inflection points (the third angular effect, Ref. 12). The transfer energies shown in Fig. 3 are consistent with previously published results.[7,12,13]

The gradual evolutions of the x-z resonances and y-z Lebed oscillations in Fig. 3 closely mimic the experimental observations. Thus, the disappearance of the x-z resonance can be regarded as neither a change in band structure nor an indication of NFL behavior (via a vanishing of t_z): *it follows directly from semiclassical Boltzmann transport* even with $\mu_0 H_y = 2.7$ T, the highest tilted field component employed in this experiment and more than ten times higher than the previously reported ''decoupling'' field of 0.2 T.

Another puzzle remains. Semiclassically, when the field is aligned along the y axis (along the open Fermi surface), one expects a nonsaturating magnetoresistance $R_{zz}(H)$. On the contrary, an initial rapid rise in resistance is followed by very weak or no magnetoresistance, at virtually all pressures. We show this in Fig. 4 for R_{zz} vs H_y at three pressures, 6.0, 7.5, and 8.3 kbar, all at $T = 0.35$ K. Again, these results are similar to what was found in $(TMTSF)_2ClO_4$, where R_{xx}, R_{yy}, and R_{zz} all tended to saturation at high field for $H \| y$ Ref. 14 (see Fig. 4 and the inset). If t_z approaches zero as in SCA, the Fermi contour washes out, so that the conductivity along that direction should vanish. This implies a diverging magnetoresistance, not the nominally field-independent behavior in Fig. 4. Could this anomalous magnetoresistance behavior still be explained by the SCA's NFL model? Once

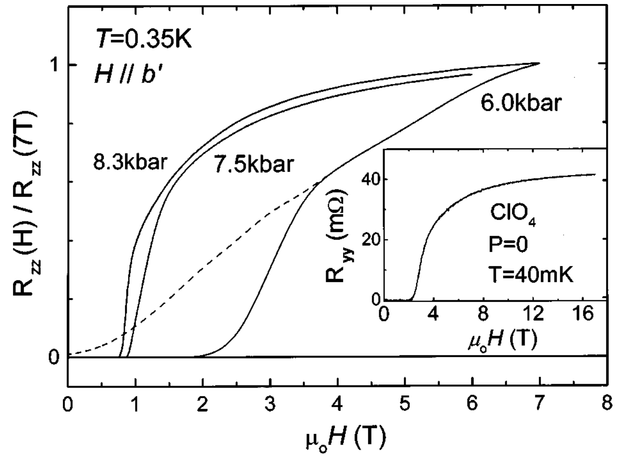

FIG. 4. Magnetoresistance at several pressures (scaled to 7-T values) for H along the open orbit y direction, showing a tendency toward saturation at high field. Semiclassics predict nonsaturating magnetoresistance in this case. The 6.0-kbar line includes a normal state extrapolation below $H_{c2} \sim 4$ T (Ref. 16). The inset shows that the intralayer resistance R_{yy}, here for the ClO_4 compound, also saturates.

the coherent interlayer transport vanishes under sufficient magnetic field H_y, a further increase of field may not affect the magnetoresistance, assuming that *field-independent incoherent* hopping is solely responsible for the finite residual conductivity. However, this seems unable to explain the fact that the saturation behavior with $H \| y$ is ubiquitous, occurring as well in the intralayer directions ρ_{xx} (Ref. 15) and ρ_{yy} (Fig. 4), where the transport always remains *coherent*. It is also noteworthy that a nontrivial temperature dependence with ''semiconducting'' behavior ($d\rho/dT<0$) has been observed in the same system at 6 kbar, in the well saturated (large H_y) regime,[16–18] rather than the metallic dependence seen at high pressure.[15,19] Therefore, the situation is rather intricate, with the saturating magnetoresistance not necessarily due to the absence of coherent transport. Different transport mechanisms may be involved at low and high pressure under large H_y. In addition, it is now known that for $H \| y$, the superconducting state is highly anomalous, with H_{c2} far exceeding the Pauli limit, likely associated with triplet pairing.[14,16] Further studies will be required to determine any relationship between the behavior in Fig. 4 and the superconducting state, where field induced dimensional crossover, but with finite interlayer transport, is predicted to lead to reentrance in high fields.[20]

In summary, we have shown that the angle dependence of the interlayer magnetoresistance in $(TMTSF)_2PF_6$ for $6.0 < P < 8.3$ kbar can be explained by semiclassical transport theory. In particular, we found from both experiment and simulation that the x-z resonance disappears smoothly under the application of a transverse magnetic field H_y (at intermediate H_y, it coexists with the Lebed y-z resonance), and that $\rho_{zz}(H_z)$ power-law scaling is absent in y-z plane rotations, with the background angular dependence becoming quite classical ($H \sin\alpha$) by 6 kbar. We also found saturating interlayer magnetoresistance with field aligned along the open Fermi surface (y axis). The first two observations suggest that, based on the SCA incoherent transport model, this system is a Fermi liquid at the pressures employed. The third point is difficult to explain from semiclassics or from a vanishing interlayer transport/decoupling model (we have noted that anomalous superconductivity was found previously in the same configuration). Taking into account previous results at ~10 kbar, this leads to the equivocal conclusion that *high* pressure drives the system into a decoupled, non-Fermi-liquid state, which should be a highly correlated system with minimal t_z. Considering the anomalous temperature dependence of the resistivity with large H_y, it is the low pressure state, close to an antiferromagnetic spin-density wave phase, that should really be considered a more likely candidate for a NFL than the high pressure state, with its larger t_z and metallic $\rho(T)$. If this is so, the T-P phase diagram would be similar to that found in the quasi-2D κ-BEDT-TTF family.[21] The question then boils down to whether or not power-law scaling of y-z (Lebed) rotations can be used as a landmark for NFL behavior in quasi-one-dimensional systems. More detailed theoretical as well as experimental work will be needed to provide a final view on this matter.

This research was supported by the National Science Foundation under Grant Nos. DMR-9258579 and DMR-9701597. We are indebted to P. M. Chaikin for use of a pressure cell and the 6.0-kbar sample used in a portion of this work.

*Present address: Department of Physics, Princeton University, Princeton, NJ 08544.

†Present address: Department of Physics, Boston College, Chestnut Hill, MA 02467. Electronic address: naughton@bc.edu

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